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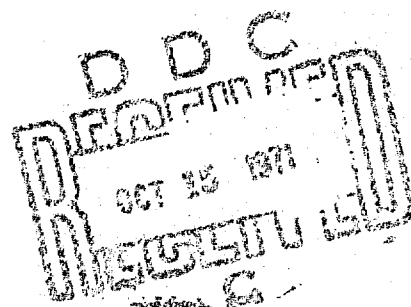
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Thermal Decomposition of Explosives

Part 8, The Reactions of Composition B and TNT
in the Presence of Various Potential Cavity Liners and Sealants

by
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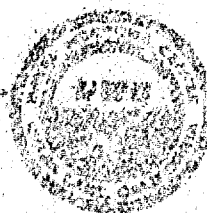
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ABSTRACT

The isothermal decompositions of Composition B and TNT were probed in the presence of a variety of materials under consideration for employment in ordnance as cavity liners or sealants. These materials included products of Essex Chemical Corp.; H. H. Robertson Co. (Tuffseal); Unique Products, Inc.; Insulation Systems Inc.; Thermal Systems, Inc.; Thiokol Chemical Corp.; and United Resin Products, Inc. The kinetic features of the reactions were established over wide temperature ranges, including conditions such that the reactions were fast and explosion imminent. In general, accelerations in the decomposition rates were observed. In a few cases these accelerations were extreme.

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AN ACTIVITY OF THE NAVAL MATERIAL COMMAND

W. J. Moran, RADM, USN Commander

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FOREWORD

The cook-off of ordnance during exposure to high temperatures—in particular, during fires—constitutes a long-standing problem. The implications for ordnance safety are obvious. Additionally, cook-off may have serious consequences from the standpoint of logistics (for instance, the loss of munitions in ammunition depot fires resulting from accident or enemy action) and shipboard operations. A particularly apparent aspect of the last is afforded by aircraft carrier operations. A desirable goal is the development of ordnance with increased resistance to accidental ignitions and greater endurance at high temperatures. The former reduces the risk of initiating fires; the latter provides additional time for countermeasures during a fire and so reduces the possibility of damaging secondary explosions.

A program at the Naval Weapons Center has been directed toward understanding the processes important to the initiation of explosions in general and the cook-off of ordnance in particular. As part of this program, a series of investigations has probed the decomposition of explosives at temperatures where reaction is rapid and explosions may occur. An important aspect of this study has been the determination of the effect of a variety of materials commonly employed in warheads on these reactions. This report presents the findings.

Part 1 presented the results of a study of the effects of an asphaltic hotmelt on the decomposition of RDX-bearing explosives. Part 2 extended the study to the reactions of this hotmelt with TNT. Part 3 investigated the decomposition of Composition B in the presence of two cavity paints. Part 4 presented a study of the reactions of RDX and Composition B in the presence of four asphalts, two waxes, and a plastisol. Part 5 extended the work of Part 4 to TNT. Part 6 investigated the decomposition of the explosive FBXW-106 and briefly studied its behavior in the presence of an asphalt. Part 7 extended the investigation of Part 6 to the explosive PBXC-11. Part 8 reports a study of the behavior of Composition B and

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TNT in the presence of eight commercial substances under consideration for use as cavity liners or sealants.

This report has been reviewed for technical accuracy by Ronald A. Henry and William P. Norris.

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INTRODUCTION

This report continues a series of investigations of the behavior of explosives at elevated temperatures where reactions become rapid and the onset of violent decomposition is imminent. The present study, conducted from April to July 1970, probed the reactions of TNT and Composition B in the presence of eight materials under consideration for use as cavity liners or sealants. Particular attention was devoted to the three substances which seemed to be of most immediate interest. The remaining materials were studied briefly, although in sufficient detail to outline their behavior over a considerable range of temperatures. The results thus provide useful information on a number of explosive/additive systems of potential practical importance. However, additional data would be necessary prior to a serious attempt to investigate fundamental reaction mechanisms.

EXPERIMENTAL

MATERIALS

The Composition B is the material used throughout this study; it is described in Part 1 (Ref. 1). The TNT is the sample recrystallized for the work of Part 5 (Ref. 2). Both explosives have been stored in opaque desiccators over calcium sulfate. The candidate liners and sealants originated from various industrial sources.^{1,2} Unfortunately, their chemical compositions are unknown. The candidates are listed and, insofar as possible, described below.

Essex Liner

Essex Liner was provided by the Essex Chemical Corp., Clifton, N.J., and identified by them as Hot Melt Coating A-968-14-52B. It is described as having a "chemically modified polyolefin" base. The brownish-gray material has the appearance and consistency of a moderately hard wax. It is readily broken or cut. In our experiments it was used in the form of fine shavings. The company reports a softening point of 143°C. Observations with a Kofler Hot Stage indicated a very slight softening at 115°C. The softening increased with rising temperature. A small amount of liquid was

¹ The author is indebted to J. M. Pakulak of NWC for the samples, for many useful discussions, and for permission to cite his work in this field.

² The citation in this report of the trade names of commercially available products does not constitute official endorsement or approval of the use of such products.

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noted at about 160°C. Although complete melting did not occur up to 230°C, increasing liquefaction and a general darkening were observed. Some slight bubbling and a considerable evolution of smoke occurred at temperatures above 210°C.

Tuffseal

Tuffseal was provided by H. H. Robertson Co., Ambridge, Pa. It is described as a two-component resin system involving petroleum asphalt, hydroxy modified polydiene, plasticizer mineral oil, alkyl tin catalyst, and nonvolatile isocyanate-terminated polymer. (The description, although not detailed, is far more complete than is usual.) Mixing the two components yielded a black, rubbery material which can be fairly easily torn apart or cut. Our experiments used fragments and chunks smaller than 1 cubic millimeter in volume. The material is described as nonmelting. Observations with a Kofler Hot Stage confirmed the statement to temperatures of at least 230°C.

Unique Liners (Hard and Soft)

Unique Products, Inc., Santa Ana, Calif., (later represented by Rocketdyne) provided two samples of a material designated as "compound 1000." These differed so greatly that it seems advisable to describe them separately. No detailed information as to the composition of the material was provided. It is formulated from a two-component system, with each component apparently containing three ingredients.

The initial sample—which will be called Hard Unique Liner—was a cured, finished product. It had the appearance of a hard, white, rather shiny, wallboard about 14 millimeters in thickness. Shavings could be obtained with a scalpel. These maintained their shape throughout the high-temperature runs, indicating an absence of melting. The second sample—called Soft Unique Liner—was mixed in the laboratory from equal quantities of two components provided by the company. Despite a reported gel time of 4 to 8 minutes (Ref. 3), the material remained fluid for more than an hour. After 15 hours it had set up to a light tan, ridged solid. The material was soft, pliable, easily cut, and had a strong solvent odor. Runs—using small chunks and shavings—were made over a period during which the material aged from 16 to 144 hours. During this interval it became somewhat less pliable and the odor lessened. The pliability gradually decreased over a period of 3 months, while the tan solid achieved a tough, hard consistency and pulled away from the watch glass on which it had been mixed. This final material was entirely different in appearance from Hard Unique Liner.³ Faint traces of odor were detectable after 5 months. No studies were made with the aged material.

Plastonium Liner

Insulation Systems Inc., Santa Ana, Calif., provided a sample—designated by them as "Plastonium material, part No. 05-69-44-01"—resembling a gray wallboard about 12 millimeters in thickness. Scraping produced a powder which was used in our studies. The material is described as

³ Recently, several samples of Unique Liner from sectioned bombs were inspected. These were similar in color and general appearance to the final Soft Unique Liner. There is no visual resemblance to Hard Unique Liner.

consisting of "several commercially available ingredients" which set up with an exothermic reaction involving a water of crystallization. The material is said to undergo phase changes at 121 and 160°C and finally convert to a "carbonaceous material."

TSI Liner

Thermal Systems, Inc., Los Angeles, provided a material designated as THERMA-LAG M-280. It is a black solid, soft enough to be indented by the fingernail and easily cut into small chunks. Its composition is unknown. It is described as consisting of a material melting at about 138°C, contained in a binder matrix melting at 232°C. Observations on the Hot Stage confirmed softening at about 138°C. The solid held its form to about 150°C; however, at this point a general breakdown commenced. Melting to a black viscous fluid was complete at 160°C. The explanation for the apparent conflict with the description provided by the company is unknown.

Thiokol Liner

Thiokol Chemical Corp., Trenton, N.J., provided a hard, reddish-brown material which does not appear to melt under the conditions of these experiments. We have no information as to its composition.

URP Liner

United Resin Products, Inc., Brooklyn, N.Y., provided a sample--designated by them as product #7126--of a translucent, white, pliable material. It is described as a "polymer resin." The company reports a melting point of 149°C. We noted that the material became transparent at about 140°C and melted to a clear liquid at about 147°C.

PROCEDURE

The equipment and general procedure have been described (Ref. 1). In brief, isothermal decompositions were carried out in a closed system under 640 torr of added nitrogen. The reaction kinetics were followed by pressure increase. The runs generally used samples of about 11 milligrams of Composition B and 20 to 25 milligrams of TNT. The liners were present in excess quantities (20 to 25 milligrams with Composition B, and 30 to 40 milligrams with TNT) to ensure adequate contact with the explosive. Blank runs were carried out with 75- to 100-milligram samples of the liners. Large gas evolutions were noted only with Plastonium and Soft Unique Liners. These will be discussed in subsequent sections. The remaining materials gave at worst (Essex and TSI Liners) gas pressures which would have been only marginally detectable under the conditions of the usual run.

Practical considerations and the large number of materials involved precluded the extensive investigations and detailed checking that would be desirable. Thus, there is a dearth of duplicate runs. Nevertheless, the compatibility of a given experiment with those at higher and lower temperatures permits considerable confidence in the data. The deviations which do appear are reported and serve

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as indicators of points which should be probed in the event that future requirements make more extensive studies of a particular liner necessary.

RESULTS

GENERAL

The current results are generally similar to the findings of the previous parts of this report. Specifically, the liners accelerated—to a greater or lesser degree—the decompositions of the explosives. The generally deceleratory reaction patterns of Composition B are conserved but with increased reaction rates. The irregular 170°C reaction (Ref. 4 and 5) is usually altered to a deceleratory pattern, although in a few cases some irregularity persists. Most of the liners convert the sigmoidal patterns of the pure TNT reaction to deceleratory decompositions, with the maximum rate at (or shortly after) initiation. The details of the reactions vary from liner to liner; hence the systems will be described separately.

Three subjects—Essex Liner, Tuffseal, and the Unique Liners—seemed to be of most immediate interest and received considerable attention. The discussion will cover first these liners with Composition B (Fig. 1-5, Tables 1-3) and with TNT (Fig. 6-10, Tables 4-7). Thereafter, the briefer investigations of the remaining liners will be summarized. Although less detailed, these were adequate to give a general picture of the behavior of the systems, indicate the temperature dependences of the reactions, and thus establish a framework upon which any necessary, future studies can be based.

THE DECOMPOSITION OF COMPOSITION B IN THE PRESENCE OF ESSEX LINER

Essex Liner very greatly accelerated the reaction of Composition B. The generally deceleratory curves are illustrated in Fig. 1 and 2, where α , the fraction of reaction as indicated by the observed pressure, is plotted against time. The smooth, deceleratory pattern persisted through a fast run at 230°C. At 240°C, however, a short burst of gas accounting for about 10% of the total gas evolution occurred at about $\alpha = 0.5$. Such bursts have been noted in the previous investigations and are presumed to signal conditions where violent explosions might be expected with larger, confined samples.

Previous reports made extensive use of the first-order decay equation

$$-\ln(1 - \alpha) = k_d t + c_0 \quad (1)$$

where t is time and k_d and c_0 are constants. The equation again proves useful in describing the Essex Liner data. An illustrative curve is presented in Fig. 3. The resulting rate constants are reported in Table 1. For comparison, the table includes data for the reaction of Composition B alone.⁴ An Arrhenius plot of the Essex Liner constants (Fig. 4) shows reasonable linearity. Some moderate

⁴ Check runs carried out throughout the course of these investigations provided considerable data on the Composition B reaction. The numbers reported in Table 1 are averages. The kinetic parameters of Table 2 differ slightly from those previously reported (Ref. 1).

scatter is heightened by the seriously low 190°C point. Previous reports have noted that reactions of complex mixtures occasionally yield anomalous runs due, probably, to poor mixing. Least-squares treatment of the Essex Liner data leads to the kinetic parameters of Table 2. Previous studies made extensive use of the reaction half-lives, $t_{1/2}$, as a simple and direct means of comparing reaction systems. Half-lives again prove useful and are reported in Table 3 and plotted in Fig. 5.

THE DECOMPOSITION OF COMPOSITION B IN THE PRESENCE OF TUFFSEAL

The reactions of Composition B in the presence of Tuffseal followed generally deceleratory patterns over the temperature range of 170 to 210°C. Short acceleratory regions did, however, appear in some reactions and gave the curves a mildly sigmoidal appearance. Figures 1 and 2 present examples. Runs at 220 and 230°C gave pressure bursts accounting for approximately 5 and 30% of the evolved gases, respectively. Such bursts have been observed fairly frequently at 240°C and more rarely at 230°C. Prior to the present study, only a single pressure burst at 220°C—caused by the very reactive Red Paint (Ref. 4)—had been noted.

Most of the Tuffseal/Composition B reaction curves are reasonably well described by Eq. 1. An example is included in Fig. 3. Rate constants are reported in Table 1. Unfortunately, the first-order plots for the 200 and 190°C runs are less successful. In these cases, as in earlier studies, two constants approximating the initial (subscript i) and final (subscript f) regions of the broken first-order plots are reported. An Arrhenius plot (Fig. 4) shows reasonable linearity and yields the kinetic parameters reported in Table 2. Half-lives are reported in Table 3 and plotted, with excellent linearity, in Fig. 5.

THE DECOMPOSITION OF COMPOSITION B IN THE PRESENCE OF HARD AND SOFT UNIQUE LINERS

Hard Unique Liner

Hard Unique Liner had relatively mild effects on the Composition B reaction. At 230°C there was a pressure burst commencing at $\alpha = 0.2$ and accounting for 50% of the evolved gases. Otherwise, moderate accelerations were observed at 210°C (Fig. 1) and 190°C. At 170°C (Fig. 2) Composition B undergoes a very drastic alteration in its reaction pattern, resulting in an initial deceleratory reaction, a slowly rising plateau, and a final acceleration and decay (Ref. 4 and 5). (A full curve for the 170°C Composition B reaction is presented in Ref. 5.) The reaction of Composition B and Hard Unique Liner (Fig. 2, Curve C) shows something of these features but with the plateau rising more steeply and accounting for the bulk of the decomposition.

The 190 and 210°C curves are fairly well described by Eq. 1 (Fig. 3, Curve C) and yield the constants reported in Table 1. Although the equation will not describe the entire 170°C curve, it will fit the initial region and give a constant representative of the first 60% of reaction. The pressure burst prevents analysis of the 220°C run. The limited data preclude any serious attempt at an Arrhenius plot. Nevertheless, the constants are displayed in Fig. 4, where a resemblance to the

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Composition B data is apparent. This may be a bit misleading. Unusual accelerations do occur, but they are of relatively short duration (see, for instance, the early region of Fig. 1, Curve C). They are thus easily elided in efforts to approximate the decomposition curve by Eq. 1 and so do not strongly influence k_d . Fortunately, they are more noticeably reflected in the reaction half-lives (Table 3). These yield a quite linear plot (Fig. 5), which can be easily compared with that of Composition B alone.

Soft Unique Liner

The Soft Unique Liner gave very different results. The runs are hard to treat, largely due to the obvious instability of the liner. The odor alone suggested volatile materials. Additionally, a 250°C blank run with 25-hour-old material gave an immediate, erratic gas evolution occurring in short bursts suggestive of eruptions through a crust. A 210°C blank run with older material (37 hours) gave a smaller and smooth gas evolution. Liner instability was also indicated by the lessening pliability of the solid during the same period. In the runs with explosives, there was evidence of an initial, anomalous gas evolution in the TNT decomposition (to be discussed subsequently) but not, curiously, with Composition B. (It should be noted that the TNT and Composition B runs were interspersed throughout the study.) Although there is some possibility that the deceleratory Composition B reaction simply masked the extraneous gas, careful study of the curves seems in fact to indicate its absence. This raises the interesting-although at present speculative-possibility of a specific interaction of the liner material with the RDX of the Composition B.

Despite the difficulties, the salient features of the Soft Unique Liner runs are clear. At 200°C and below there were obvious-but not extreme-increases in reaction rates relative to those produced by Hard Unique Liner. The seriously disturbing problem occurred at 210°C, where a run with 21-hour-old liner resulted in an explosion after 104 seconds. The explosion differed from the usual pressure bursts. The latter consume only a portion of the sample, leaving the remainder to follow a smooth decomposition. The explosion with Soft Unique Liner was to completion. As the study progressed and it became obvious that the properties of the liner were changing, it seemed possible that older material would give less violent reactions. (The supposition was reinforced by the rather leisurely 200°C run with 67-hour-old material.) Therefore, a second 210°C run with 89-hour-old material was attempted. In this case an explosion-again to completion-occurred at 138 seconds.

The obvious instability of the liner and consequent irreproducibility in the system makes detailed analysis of the rate data relatively useless. Nevertheless, rate constants and half-lives were obtained and are included in the tables for purposes of record and comparison with the Hard Unique Liner. There is clearly little point in attempting to obtain kinetic parameters for a system of this nature.

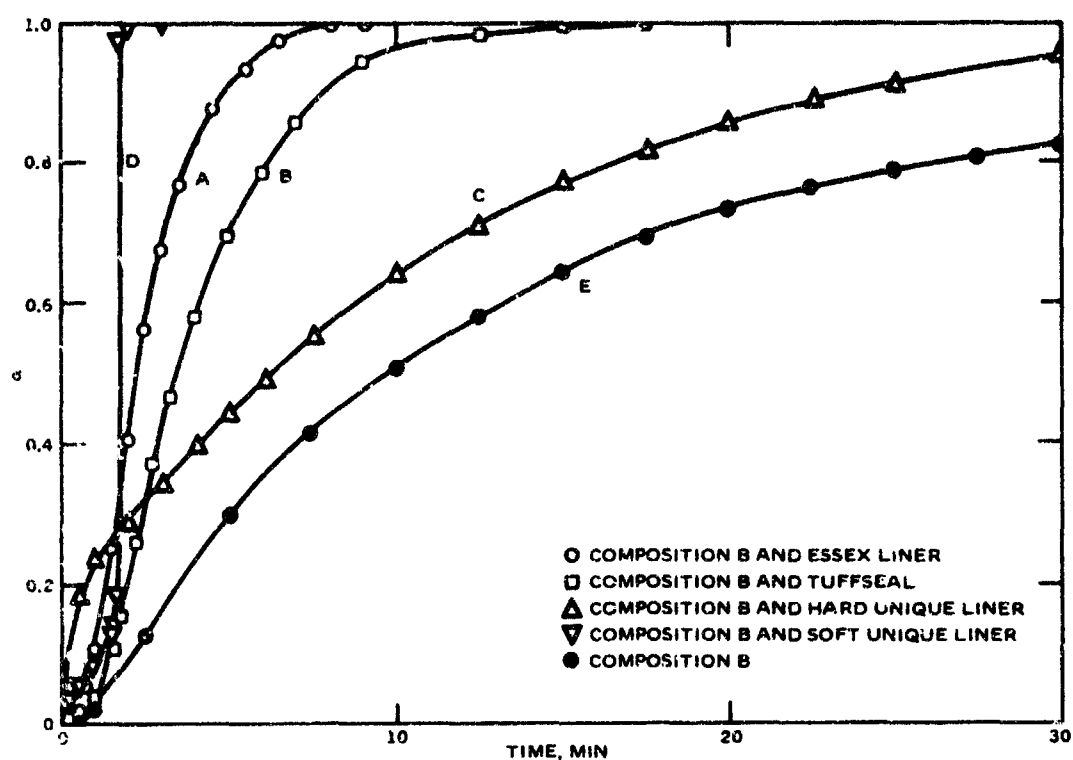


FIG. 1. Thermal Decomposition of Composition B at 210°C and in the Presence of Various Materials.

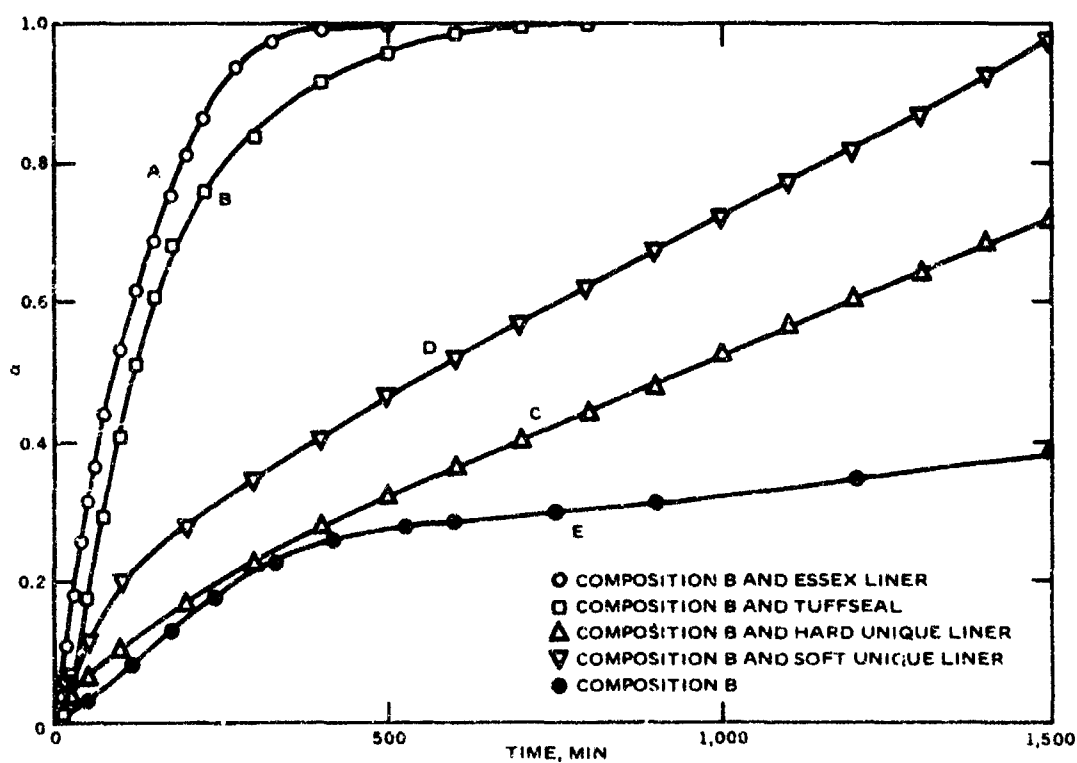


FIG. 2. Thermal Decomposition of Composition B at 170°C and in the Presence of Various Materials.

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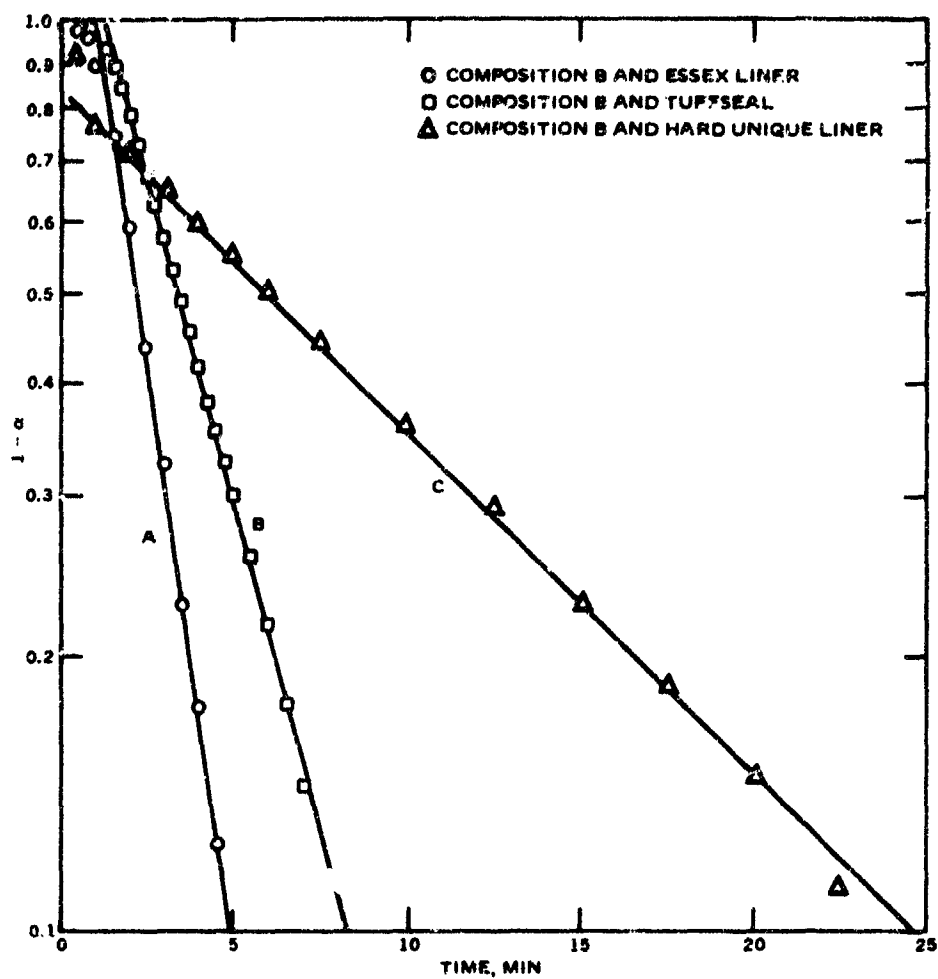


FIG. 3. Illustrative Curves for the Treatment of the Composition B/Liner Reactions by Eq. 1. Temperature : 210°C.

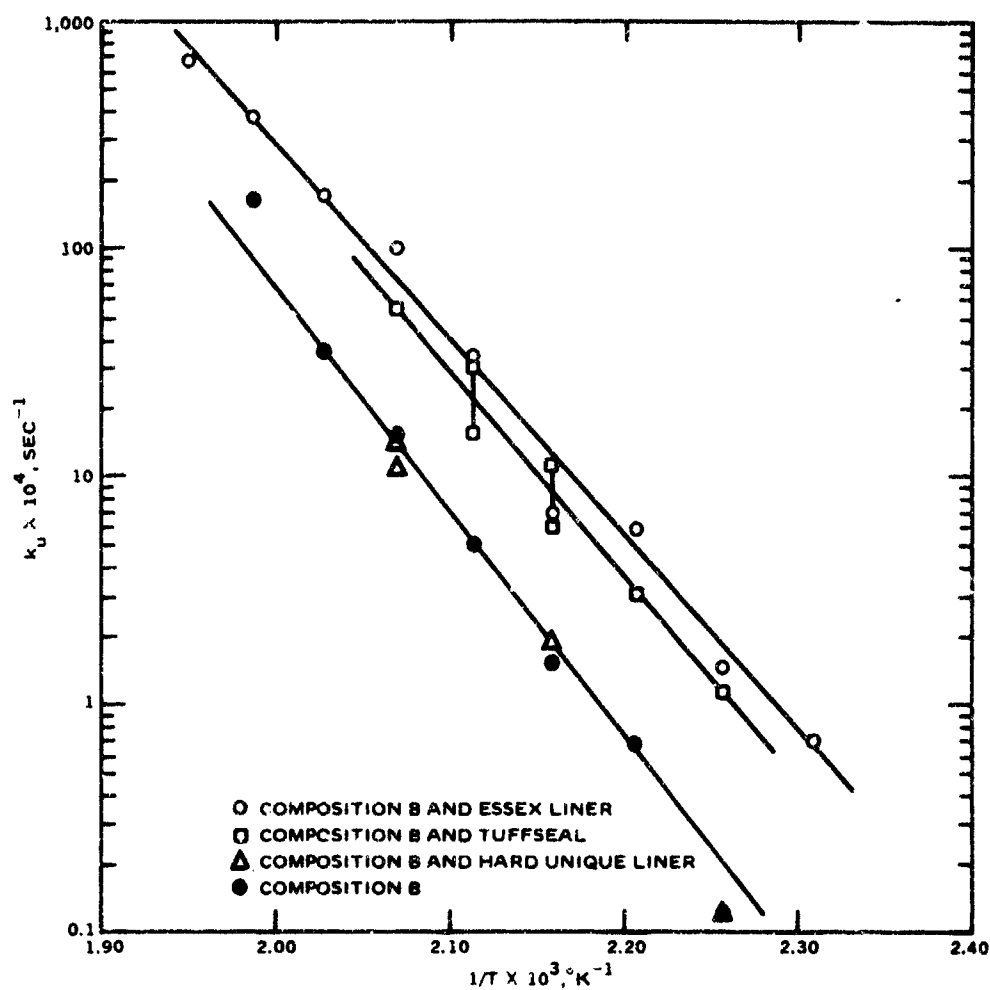


FIG. 4. Arrhenius Plots for the Decomposition of Composition B in the Presence of Various Materials. Bars connect the constants from runs giving broken first-order plots.

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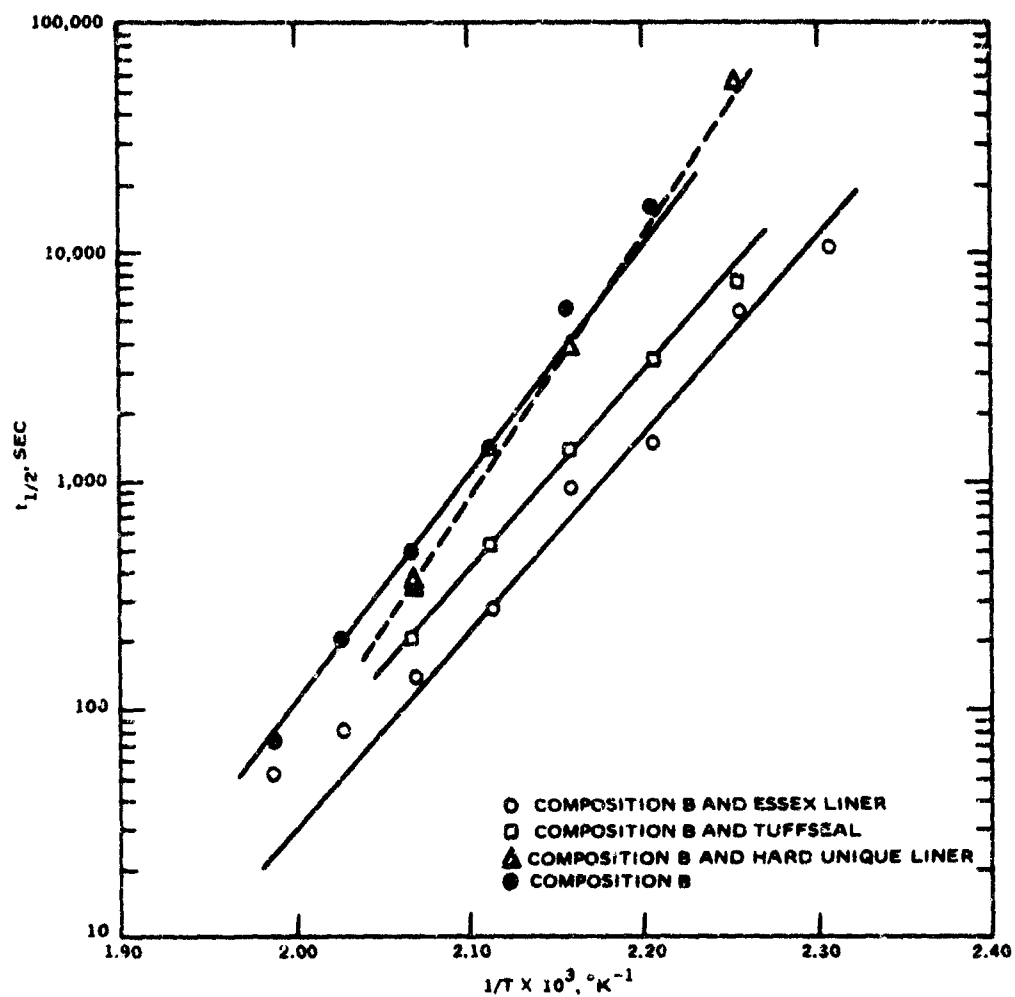


FIG. 5. Half-Life Plots for the Decomposition of Composition B in the Presence of Various Materials. The half-life for the 170°C Composition B run (142,200 seconds) falls off scale.

TABLE 1. Rate Constants (k_u) for the Decomposition of Composition B in the Presence of Essex Liner, Tuffseal, and Unique Liners.

Temperature, °C	$k_u \times 10^4, \text{sec}^{-1}$				
	Additive				
	None ^a	Essex Liner	Tuffseal	Hard Unique Liner	Soft Unique Liner
240	...	675 ^b
230	162	383
220	35.7	172
210	14.7	100	55.2	11.4 ^d	...
				14.1 ^d	...
200	5.14	33.5	15.7 _i ^e	...	4.49
			30.6 _f
190	1.56	6.98	6.06 _i ^e	1.85	2.51
			11.2 _f
180	0.671	5.89	3.05
170	0.121 ^f	1.43	1.12	0.123 ^f	2.14
160	...	0.687

^a The rate constants for the reaction of Composition B alone are average values.

^b This run showed a short burst at $\alpha = 0.5$. The constant describes the final 40% of reaction.

^c Pressure burst prevented the estimation of rate constants.

^d Duplicate runs.

^e Constants from broken curves yielding initial (subscript i) and final (subscript f) linear regions.

^f The rate constants describe the early regions of runs having the irregular character of the 170°C Composition B reaction (Ref. 4 and 5).

TABLE 2. Apparent Activation Energies (E_a) and Preexponentials (A) for the Decomposition of Composition B in the Presence of Tuffseal and Essex Liners.

Additive	$E_a, \text{kcal/mole}$	$\text{Log } A, \text{sec}^{-1}$
None	45.3 ^a	17.62 ^a
Essex Liner	39.7	15.79
Tuffseal	41.2	16.37

^a Least-squares fit of the 180 to 220°C data. Including the fast 230°C run and the constant from the irregular 170°C curve steepens the Arrhenius plot and yields $E_a = 50.4 \text{ kcal/mole}$ and $\text{log } A = 19.97$.

TABLE 3. Half-Lives ($t_{1/2}$) for the Decomposition of Composition B in the Presence of Essex Liner, Tuffseal, and Unique Liners.

Temperature, °C	$t_{1/2}$, sec ^a				
	Additive				
	None ^b	Essex Liner	Tuffseal	Hard Unique Liner	Soft Unique Liner
240	...	33 ^c
230	73	53	54 ^c	47 ^c	...
220	203	81	81 ^d
210	490	139	204	348 ^e	104 ^{c,e}
				378 ^e	138 ^{c,e}
200	1,428	275	522	...	1,188
190	5,680	936	1,374	3,780	3,288
180	15,900	1,452	3,360
170	142,200	5,460	7,260	55,800	33,720
160	...	10,020

^a The reported half-lives are the actually observed times without correction for a warm-up period of about 30 seconds.

^b Average values.

^c Achieved during a burst.

^d Achieved after a short burst.

^e Duplicate runs.

THE DECOMPOSITION OF TNT IN THE PRESENCE OF ESSEX LINER

Essex Liner produced very serious accelerations in the TNT decompositions. The severity of the effect is heightened by the elimination of the early, relatively slow reaction characteristic of pure TNT. Instead, generally deceleratory reactions achieve maximum rates very shortly after initiation. Figures 6 and 7 present illustrative curves. In general, the various TNT/liner curves are less well described by Eq. 1 than was the case with Composition B. Broken first-order plots are common. Figure 8 is illustrative. In these cases, reasonable approximations are usually given by a pair of straight lines. Rate constants are presented in Table 4. The Arrhenius plot (Fig. 9) for TNT and Essex Liner shows a rather considerable scatter. As drawn, the line stresses the linear 220 to 260°C data and gives a reasonable indication of the temperature dependence at lower temperatures. The resulting kinetic parameters are reported in Table 5. Although direct comparison of rate constants from deceleratory and sigmoidal curves is impossible, the reaction half-lives (Table 6) provide a clear illustration of the differences in the decomposition of pure TNT and TNT in the presence of Essex Liner. The very large accelerations are even more apparent in the half-life plots (Fig. 10).

THE DECOMPOSITION OF TNT IN THE PRESENCE OF TUFFSEAL

Tuffseal also converted the TNT reaction to generally deceleratory patterns, although short acceleratory regions gave a few curves mildly sigmoidal appearances. The rate increases are extreme. The decompositions are even faster than those with Essex Liner. Figures 6 and 7 present illustrative curves. The reaction becomes fast at 230°C, and a short burst accounting for about 15% of the gas evolution is observed at 240°C. These are much lower temperatures—by some 60°C—than would be required for comparable rates with pure TNT. The low-temperature burst is also very uncharacteristic of TNT reactions. Despite the slightly sigmoidal appearance of some reaction curves, descriptions by Eq. 1 are possible (Fig. 8, Curve B). An Arrhenius plot shows reasonable linearity (Fig. 9) and least-squares treatment of the data (excluding the fast 230°C run) yields the kinetic parameters reported in Table 5. The half-life plot (Fig. 10) also shows good linearity.

It should be noted that the Tuffseal/TNT gas evolution is somewhat low—about 60 to 70% that of pure TNT and other TNT/liner reactions—at temperatures below 220°C. At 230 and 240°C it is approximately normal. Although the explanation for the decrease is unknown, it should be emphasized that obvious reactions occurring at rapid rates give clear indications of potential trouble in practical applications.

THE DECOMPOSITION OF TNT IN THE PRESENCE OF HARD AND SOFT UNIQUE LINERS

Hard Unique Liner

Hard Unique Liner produces far greater accelerations with TNT than with Composition B. The sigmoidal character of the TNT curve is not entirely destroyed; however, the reaction rates are very greatly increased. Figures 6 and 7 present illustrative curves. Equation 1 has only moderate success with the higher temperature runs (Fig. 8, Curve C) and fails badly at 190°C. Although the resulting rate constants are reported in Table 4 and displayed in Fig. 9, they are too few and scattered for an Arrhenius plot. Previous analyses of sigmoidal TNT curves (Ref. 6) used the very simple approximation of a linear rate law,

$$\alpha = k_1 t + c_1 \quad (2)$$

In the case of TNT and Hard Unique Liner, Eq. 2 describes the region $0.2 < \alpha < 0.8$ reasonably well. The resulting constants (Table 7) yield a fairly linear Arrhenius plot (Fig. 9) and the kinetic parameters reported in Table 5. The half-life plot is linear for the 190 to 230°C runs (Fig. 10), with the 250°C point falling rather high. Some of the difficulties with these runs, and a certain inconsistency from one treatment to another, may be due to poor contact between the liner shavings and the explosive. For instance, a delay in the molten TNT's establishing complete contact with the shavings could give an overly long $t_{1/2}$, as observed at 250°C, whereas the subsequent decomposition (with good contact) could proceed at the expected speed and give a linear rate constant compatible with the lower temperature data.

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Soft Unique Liner

Soft Unique Liner presents problems—touched on earlier—which hinder sensible analysis. The liner is unstable. Moreover, with TNT there is an initial, fast gas evolution followed by partial adsorption or condensation. This leads to a spike in the pressure trace and raises obvious analytical problems. Specifically, the early gas can be considered a part of the reaction, and α can be calculated on the basis of the final, observed pressure. Figure 6 presents a curve of this type. Alternatively, an approximate correction for the early gas can be made so that α is based, presumably, on only the gas evolved by the decomposing explosive. Both treatments have the obvious drawback of assuming far too much about the chemical and physical behavior of the initial gas. Nevertheless, constants and half-lives based on both approaches are reported in Tables 4 and 6. These should be taken as only rough measures of the behavior of the system. Their principal value lies in the comparison they permit with the Hard Unique Liner data. Once again, the instability of the system prevents the determination of useful kinetic parameters.

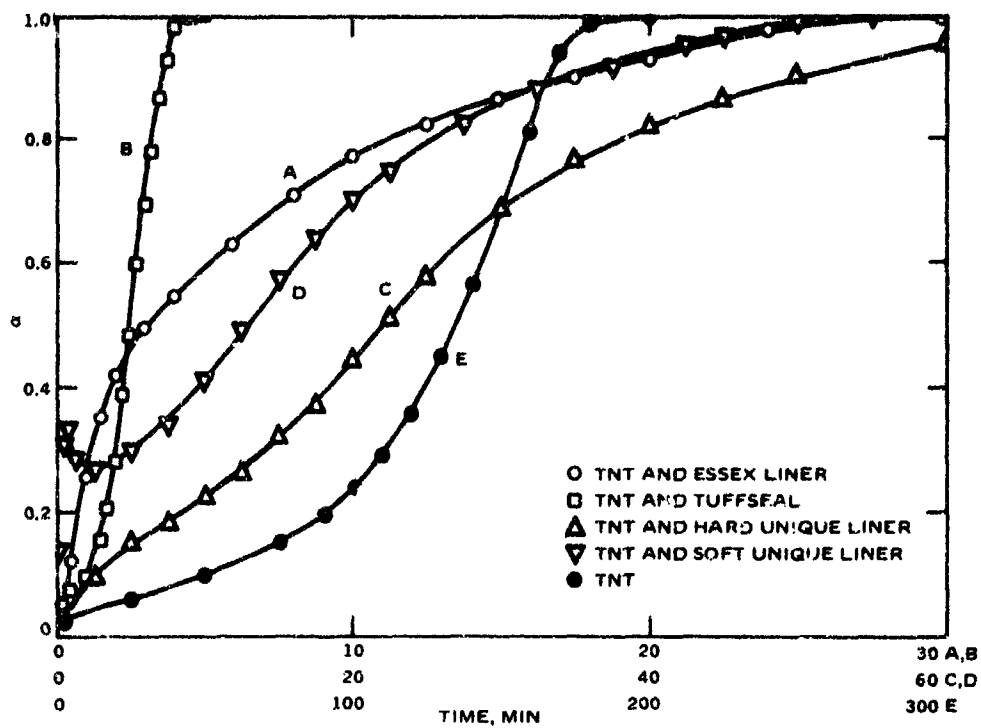


FIG. 6. Thermal Decomposition of TNT at 250°C and in the Presence of Various Materials.

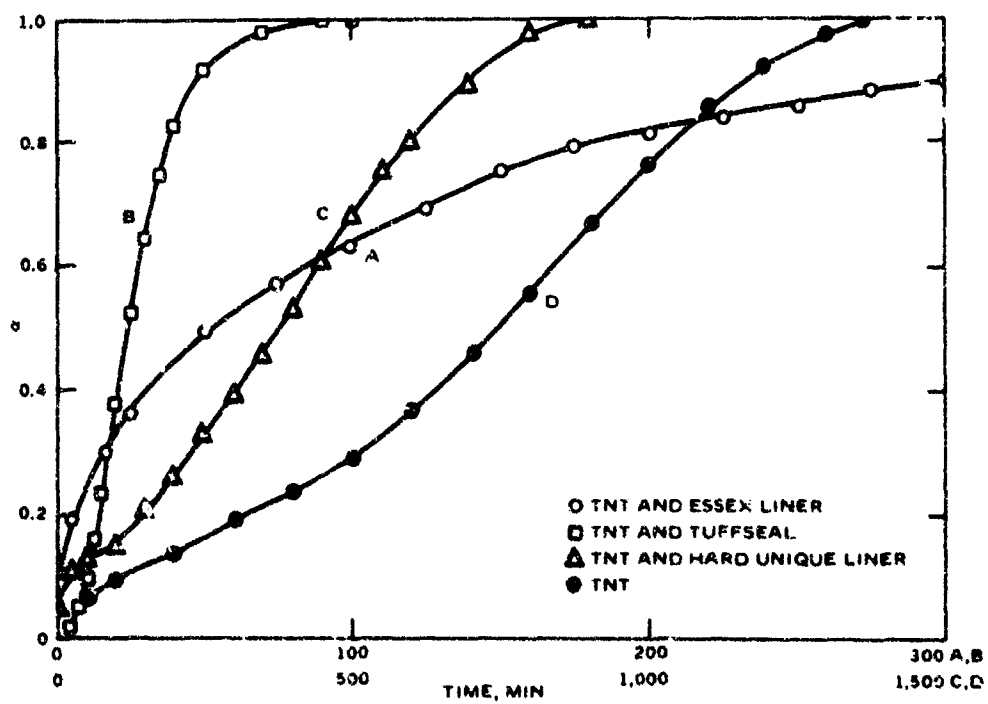


FIG. 7. Thermal Decomposition of TNT at 190°C and in the Presence of Various Materials.

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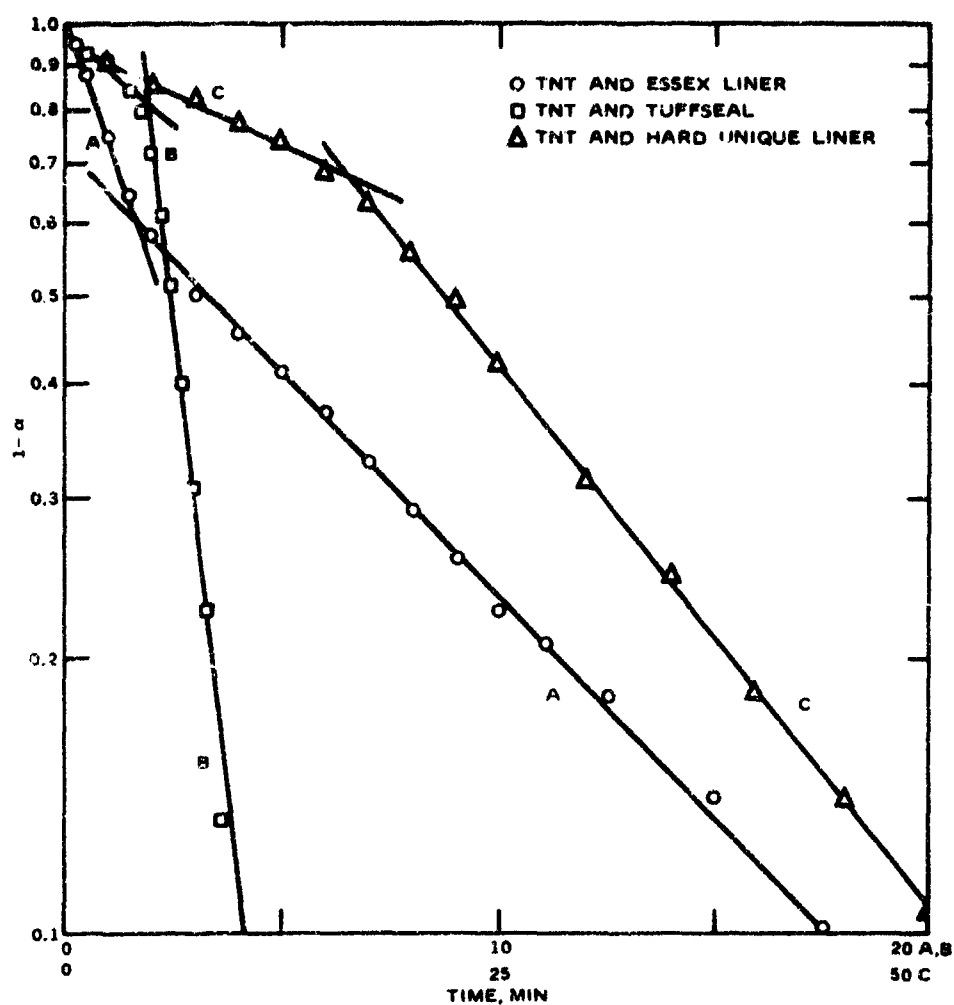


FIG. 8. Illustrative Curves for the Treatment of TNT/Liner Reactions by Eq. 1.
Temperature: 230°C.

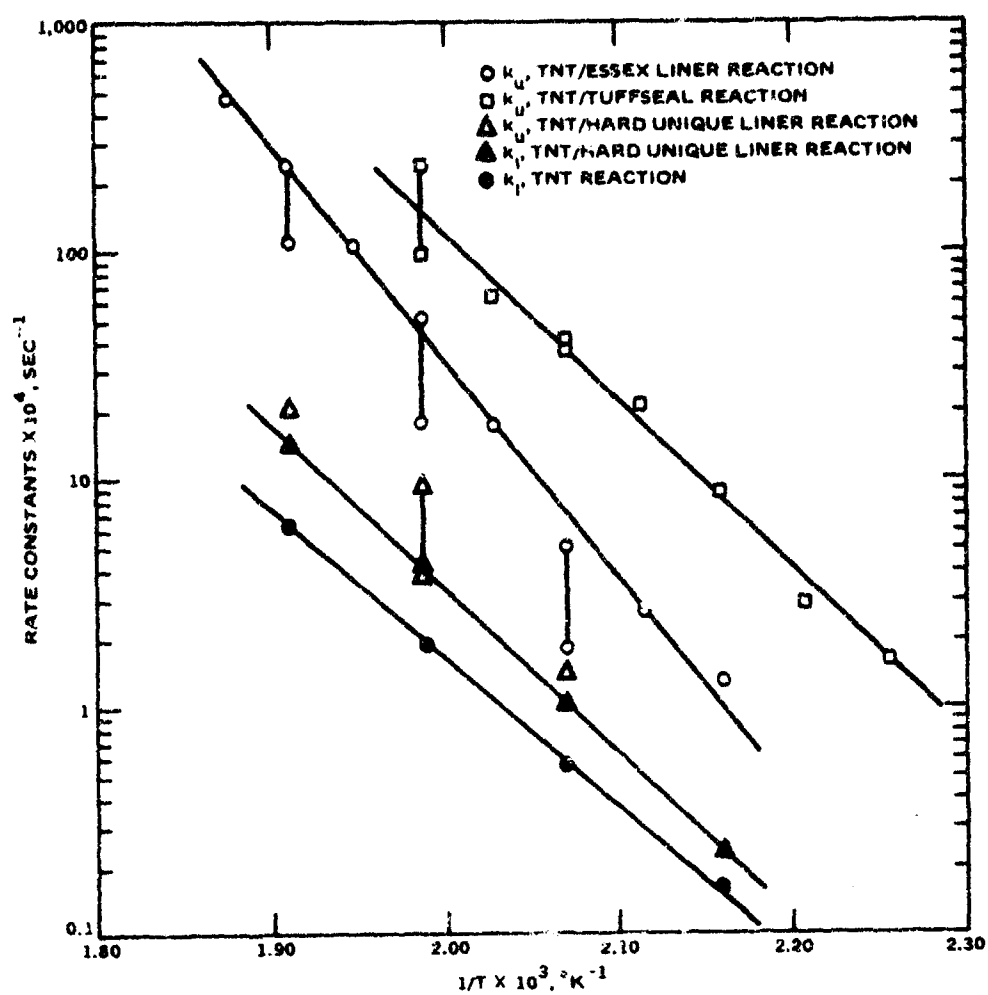


FIG. 9. Arrhenius Plots for the Decomposition of TNT in the Presence of Various Materials. Bars connect the constants from runs giving broken first-order plots.

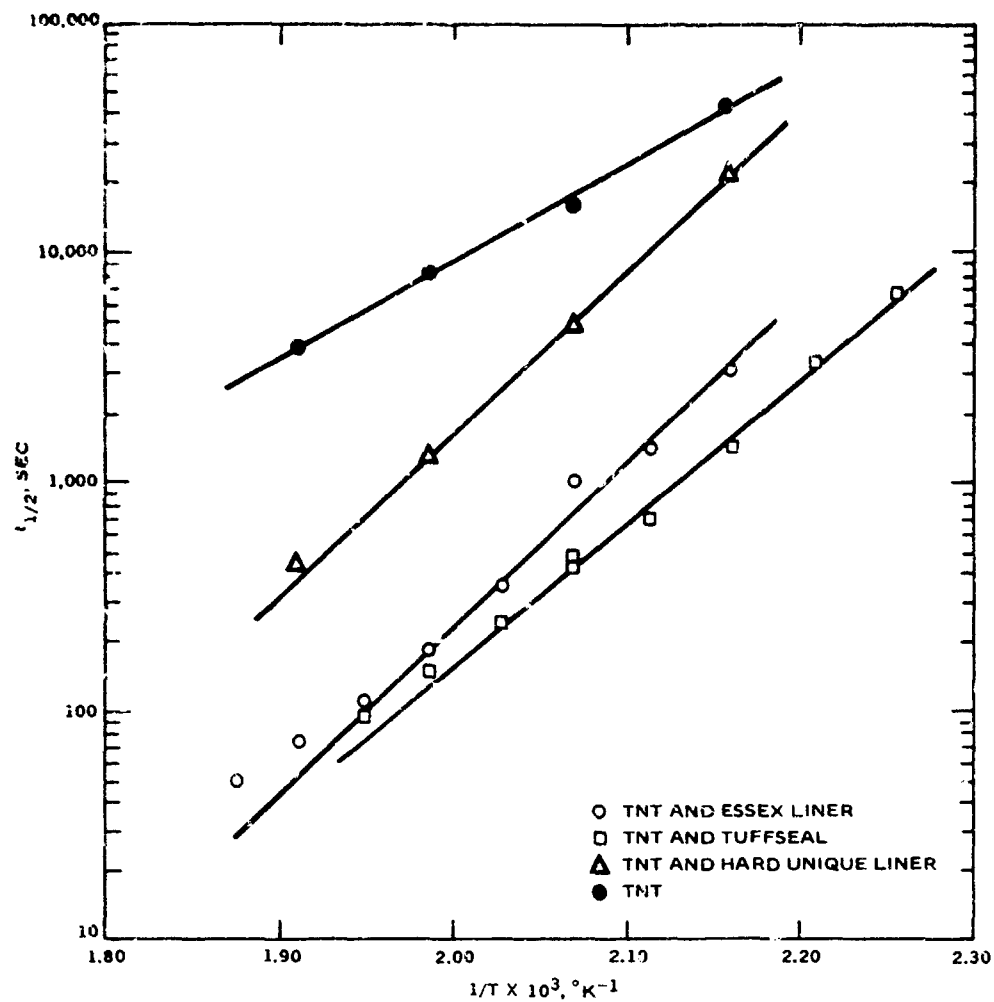


FIG. 10. Half-Life Plots for the Decomposition of TNT in the Presence of Various Materials.

TABLE 4. Rate Constants (k_u) for the Decomposition of TNT in the Presence of Essex, Tuffseal, and Unique Liners.

Temperature, °C	$k_u \times 10^4, \text{sec}^{-1}$				
	Additive				
	Essex Liner	Tuffseal	Hard Unique Liner	Soft Unique Liner	
				Uncorrected	Corrected ^d
260	486
250	111 _i ^b 243 _f	...	20.9	53.0	55.8
240	107
230	51.7 _i ^b 17.9 _f	98.8 _i ^b 244 _f	3.84 _i ^b 9.45 _f	11.6	11.9
220	17.2	64.2
210	5.05 _i ^b 1.83 _f	38.0 40.8 ^d	1.41	3.16	3.28
200	2.62	21.3
190	1.30	8.93	...	0.560	0.568
180	...	2.89
170	...	1.61

^a The correction is for an early, anomalous gas evolution.

^b Constants from broken curves yielding initial (subscript i) and final (subscript f) linear regions.

^c A pressure burst prevented estimation of a rate constant.

^d This run used a large (42.9-mg) sample of TNT.

^e The run could not be approximated by Eq. 1.

TABLE 5. Apparent Activation Energies (E_a) and Preexponentials (A) for the Decomposition of TNT in the Presence of Essex Liner, Tuffseal, and Hard Unique Liners.

Additive	Treatment	E_a , kcal/mole	Log A, sec ⁻¹
None ^a	Eq. 2	30.1	9.37
Essex Liner	Eq. 1	43.9	16.71
Tuffseal	Eq. 1	33.8	12.84
Hard Unique Liner	Eq. 2	33.3	11.06

^a The TNT decomposition is more fully discussed in the section dealing with the Plastonium and URP Liner studies.

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TABLE 6. Half-Lives ($t_{1/2}$) for the Decomposition of TNT in the Presence of Essex Liner, Tuffseal, and Unique Liners.

Temperature, °C	$t_{1/2}$, sec ^a					
	Additive					
	None ^b	Essex Liner	Tuffseal	Hard Unique Liner	Soft Unique Liner	
					Uncorrected	Corrected
260	...	50
250	3,978	72	...	438	265	315
240	...	103	100 ^c
230	8,064	181	151	1,308	762	1,002
220	...	351	239
210	16,080	990	449	4,800	1,908	2,580
			469 ^d			
200	...	1,350	678
190	44,340	3,060	1,446	22,680	9,180	12,300
180	3,450
170	6,780

^a The reported half-lives are uncorrected for a warm-up time of about 30 seconds.

^b The data were derived from calibration runs with the TNT sample used in this study.

^c Observed prior to a short burst.

^d This run used a large (42.9-mg) sample of TNT.

TABLE 7. Rate Constants (k_1) for the Decomposition of TNT in the Presence of Hard Unique Liner.

Temperature, °C	$k_1 \times 10^4$, sec ⁻¹	
	TNT ^a	TNT and Hard Unique Liner
250	6.28	14.3
230	1.95	4.28
210	0.551	1.01
190	0.161	0.227

^a The linear approximation to the TNT decompositions is more fully discussed in the section dealing with Plastonium and URP Liners.

THE DECOMPOSITION OF COMPOSITION B IN THE PRESENCE OF TSI, THIOKOL, PLASTONIUM, AND URP LINERS

The briefer studies of the reactions of Composition B and the remaining four liners are illustrated in Fig. 11 and 12. The characters of the decompositions are quite varied. TSI Liner yields generally deceleratory curves and accelerations similar to those produced by asphalts. Thiokol Liner has a less drastic effect, although accelerations are still observed. At 170°C (Fig. 12, Curve B) Thiokol Liner only partially removes the irregular character of the low-temperature Composition B reaction. URP Liner is, overall, the most inert material discussed in this report. It produces only minor accelerations at high temperatures. At 170°C the decomposition curve retains the irregular character of the Composition B reaction despite a notable acceleration in the later stages of the run.

Plastonium Liner also has mild effects on the explosive but presents the special problem of a very rapid evolution of water. (It seems reasonable to relate this to the water of hydration mentioned in the description of Plastonium.) This causes pressure traces qualitatively similar to those of TNT and Soft Unique Liner—including an early spike—and presents the same dilemma for analysis. In the present case, the clearly extraneous water has been corrected for. The reaction curves (Fig. 11 and 12) are thus based on the gas evolved by the decomposing explosive. The correction is only approximate, and the apparent slight inhibition at 210°C (Fig. 11) probably reflects its inadequacy. At lower temperatures (Fig. 12) an obvious acceleration of the irregular Composition B curve is apparent.

Equation 1 is reasonably descriptive of the various reaction curves. The resulting constants are reported in Table 8. Although too sparse for confident Arrhenius plotting, the constants are displayed in Fig. 13 and compared with the results for Composition B alone. The accelerations caused by TSI Liner are apparent. The Thiokol Liner constants reflect the initial rapidity of the decomposition and the subsequent slowing to a rate paralleling the Composition B reaction. The constants for the URP Liner and corrected Plastonium reactions show close agreement with the Composition B data. Unfortunately, at 170°C this reflects, primarily, the stress the treatment places on the early regions of the irregular curves. The rate constants should, therefore, be balanced out against the reaction half-lives (Table 9 and Fig. 14), which give an indication of the low-temperature irregularities.

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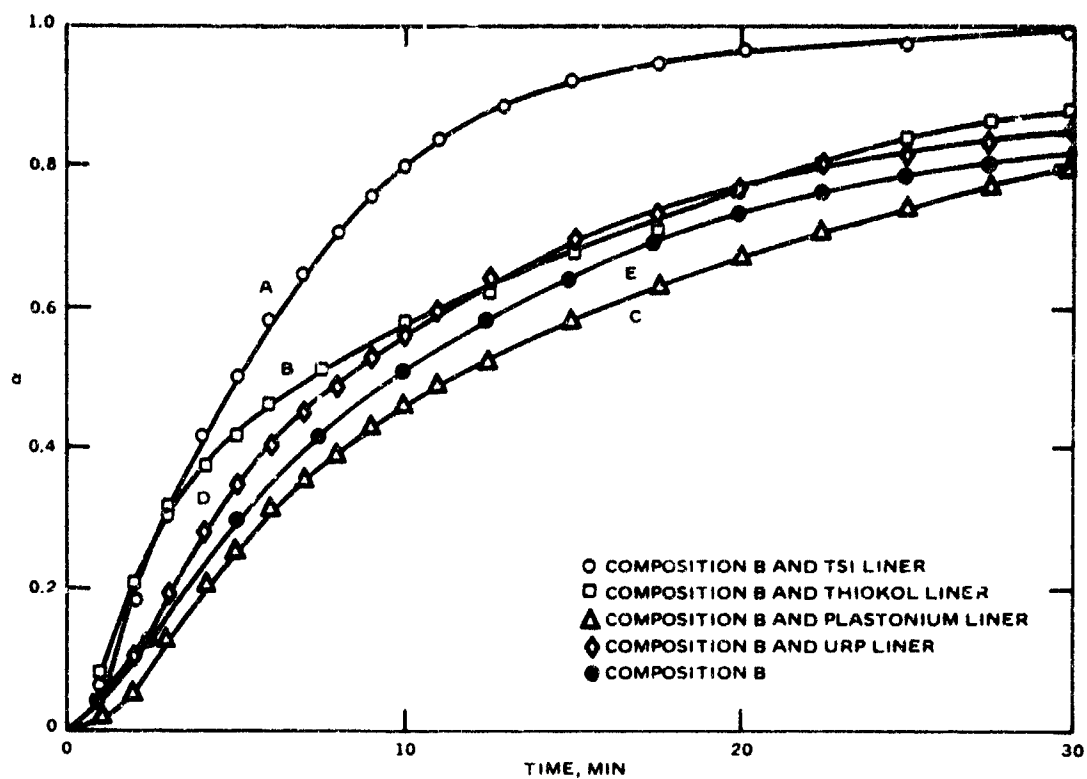


FIG. 11. Thermal Decomposition of Composition B at 210°C and in the Presence of Various Materials.

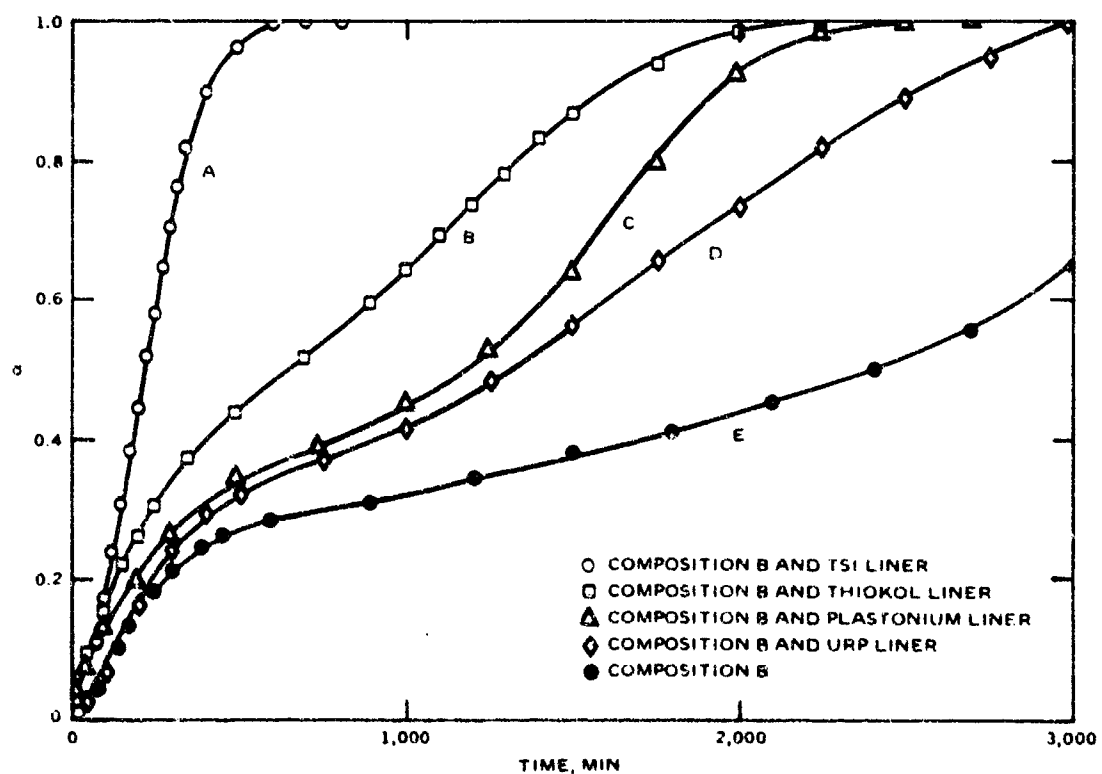


FIG. 12. Thermal Decomposition of Composition B at 170°C and in the Presence of Various Materials.

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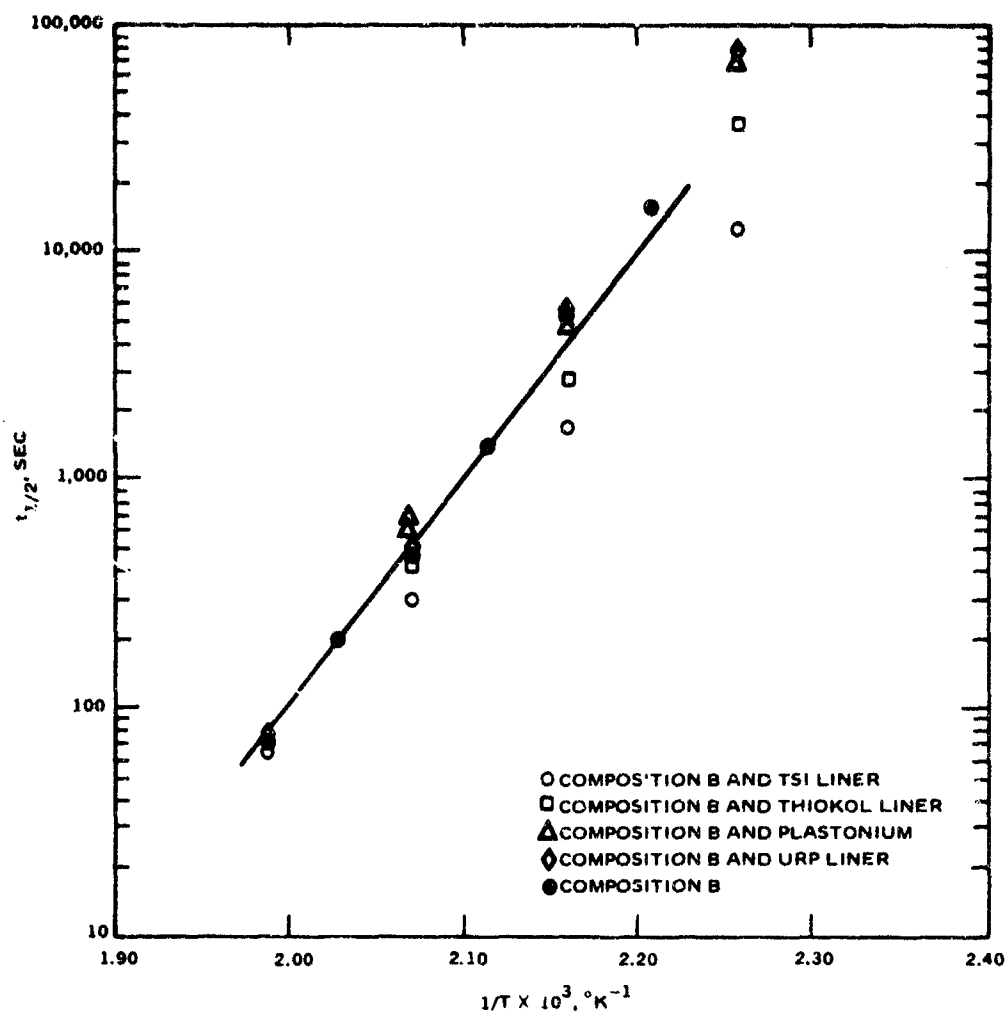


FIG. 14. Half-Life Plots for the Decomposition of Composition B in the Presence of Various Materials. The line shown has a slope appropriate for the 45.3 kcal/mole apparent activation energy of the Composition B reaction.

TABLE 8. Rate Constants (k_u) for the Decomposition of Composition B in the Presence of TSI, Thiokol, Plastonium, and URP Liners.

Temperature, °C	$k_u \times 10^4, \text{sec}^{-1}$				
	Additive				
	None	TSI Liner	Thiokol Liner	Plastonium ^d Liner	URP Liner
230	162	177	... ^b	... ^b	149
210	14.7	27.8	24.1 _i ^c 10.5 _f	9.55 ^d 11.4 ^d	14.9
190	1.56	4.67 _i ^c 7.97 _f	2.77 _i ^c 1.79 _f	1.59	1.41
170	0.121 ^e	0.594 _i ^c 1.48 _f	0.235 ^e	0.238 ^e	0.174 ^e

^a The rate constants for the Plastonium Liner are based on data corrected for an initial evolution of water.

^b Pressure bursts prevented the estimation of rate constants.

^c Constants from broken curves yielding initial (subscript i) and final (subscript f) linear regions.

^d Duplicate runs.

^e The rate constants describe the early regions of runs having the irregular character of the 170°C Composition B reaction (Ref. 4 and 5).

TABLE 9. Half-Lives ($t_{1/2}$) for the Decomposition of Composition B in the Presence of TSI, Thiokol, Plastonium, and URP Liners.

Temperature, °C	$t_{1/2}, \text{sec}^d$				
	Additive				
	None	TSI Liner	Thiokol Liner	Plastonium ^b Liner	URP Liner
230	73	66	52 ^c	46 ^c	77
210	400	299	426	672 ^d 678 ^d	492
190	5,680	1,728	2,880	5,130	5,400
170	142,200	13,020	38,640	70,320	78,300

^a The reported half-lives are uncorrected for a warm-up time of about 30 seconds.

^b The Plastonium Liner half-lives are based on data corrected for an initial evolution of water.

^c Attained during a burst.

^d Duplicate runs.

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THE DECOMPOSITION OF TNT IN THE PRESENCE OF TSI, THIOKOL, PLASTONIUM, AND URP LINERS

The decompositions of TNT in the presence of TSI, Thiokol, Plastonium, and URP Liners are illustrated in Fig. 15 and 16. All of the liners accelerate the reaction. TSI and Thiokol Liners have the greatest effects. The generally deceleratory TSI and Thiokol Liner curves are quite well described by Eq. 1. Rate constants are reported in Table 10. The reactions with Plastonium and URP Liners retain sigmoidal curves. There is, however, a marked shortening of the early acceleratory periods. (The Plastonium curve is again corrected for the evolution of water.) Equation 1 cannot, of course, describe the entire course of sigmoidal curves. The previous study of TNT (Ref. 6) used both Eq. 2 and Robertson's (Ref. 7) logarithmic equation

$$\log_{10} \alpha = k_a t + c_a \quad (3)$$

Both treatments have been applied to the curves for pure TNT and TNT with Plastonium and URP Liners. The linear equation was generally successful for the regions $0.30 < \alpha < 0.85$. Rate constants are reported in Table 11. Equation 3 was less successful with the TNT/liner systems but gave good descriptions of the TNT runs (Fig. 17). Rate constants are given in Table 12. In the case of the TNT/liner runs these approximately describe the regions $0.1 < \alpha < 0.5$.

Once again the data are too few for reliable Arrhenius plotting. Figure 18 displays the first-order rate constants for the TSI and Thiokol Liner reactions and the linear rate constants from the Plastonium and URP Liners and pure TNT reactions. The most obvious feature of the plots is the close paralleling of the TNT/liner curves to the linear rate constants for pure TNT. The results of the rather unsuccessful analyses by Eq. 3 are not displayed. The k_a 's for pure TNT gave a disappointing Arrhenius plot in spite of the linearity of the individual $\log \alpha$ versus time plots. Conversely, the TNT/liner Arrhenius plots were fairly linear; however, since the rate constants are not overly descriptive (Fig. 18), it is best to report the treatment as unsatisfactory insofar as the present data are concerned.

The half-lives (Table 13) yield fairly linear plots (Fig. 19). Again, the number of data points is limited. Nevertheless, comparison with the line for pure TNT emphasizes the decreases in reaction times brought about by the additives.

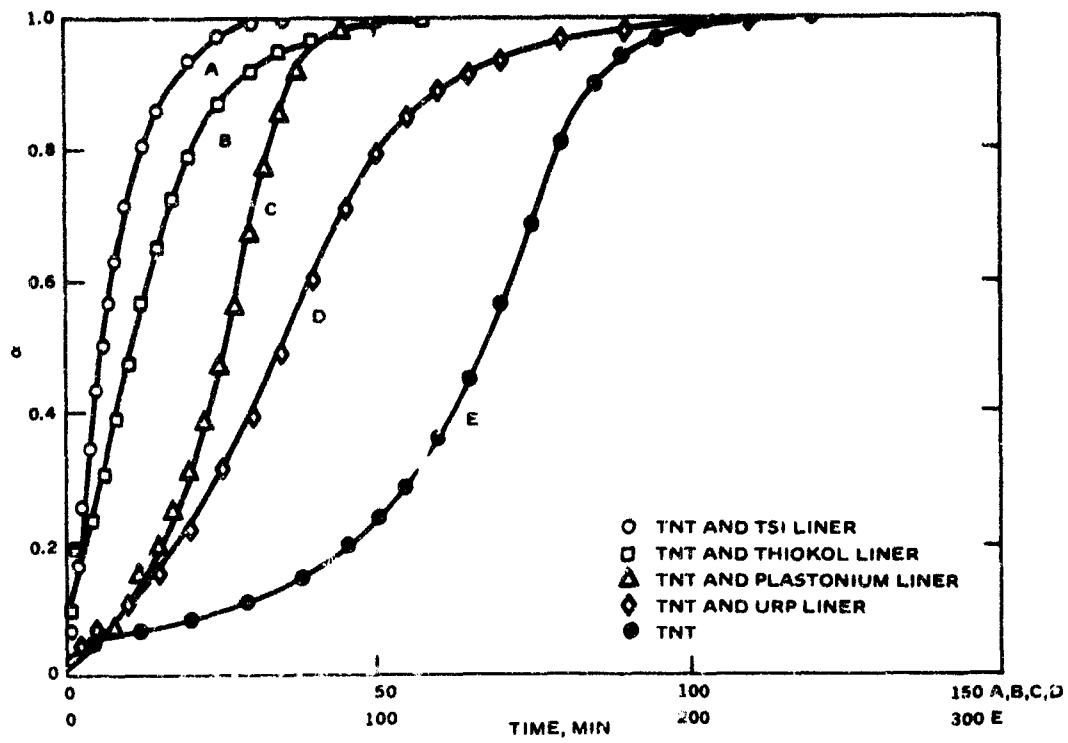


FIG. 15. Thermal Decomposition of TNT at 230°C and in the Presence of Various Materials.

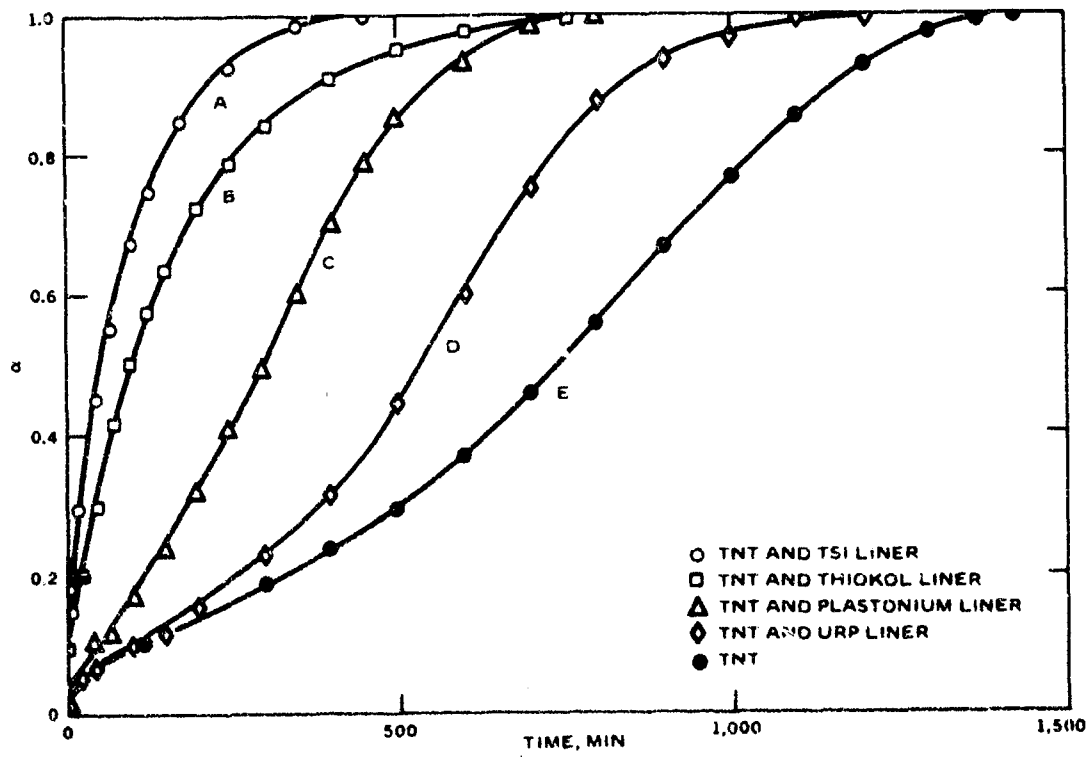


FIG. 16. Thermal Decomposition of TNT at 190°C and in the Presence of Various Materials.

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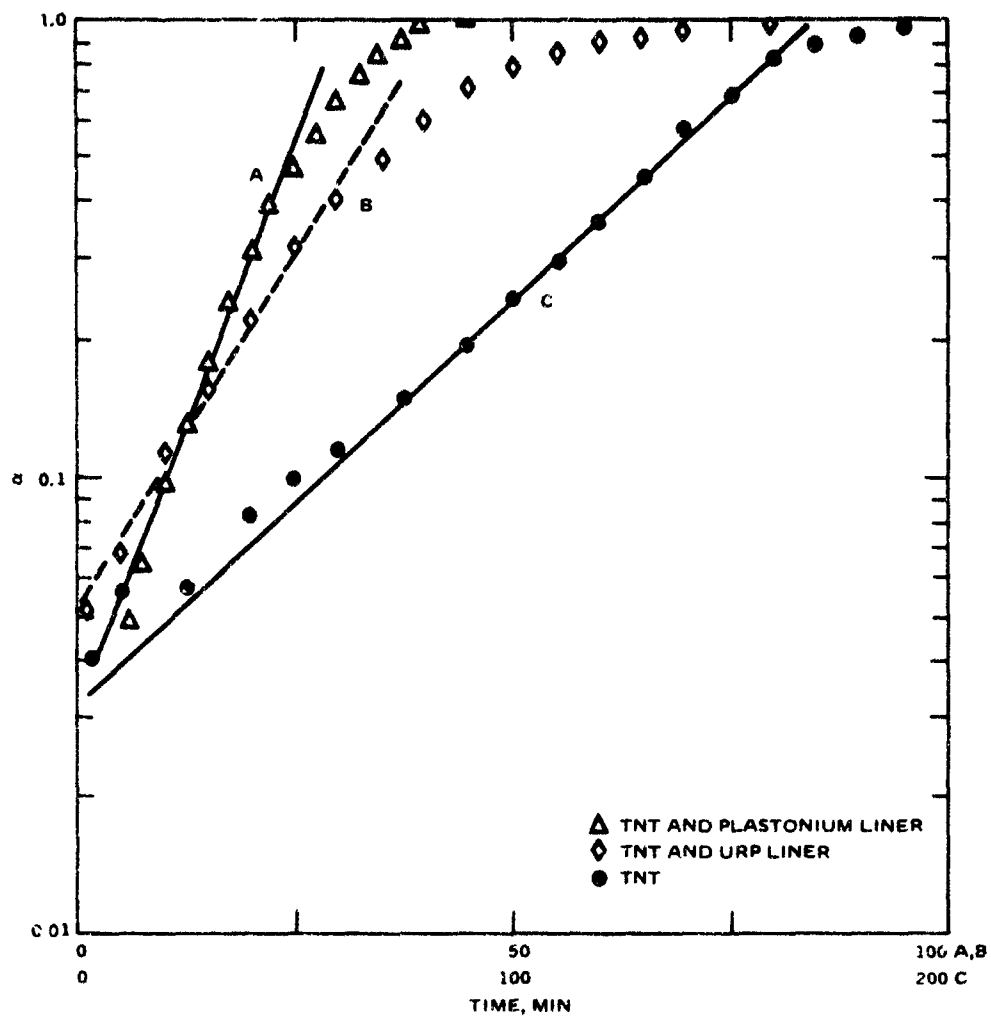


FIG. 17. Illustrative Curves for the Treatment of the TNT/Liner Reactions by Eq. 3. Temperature: 230°C.

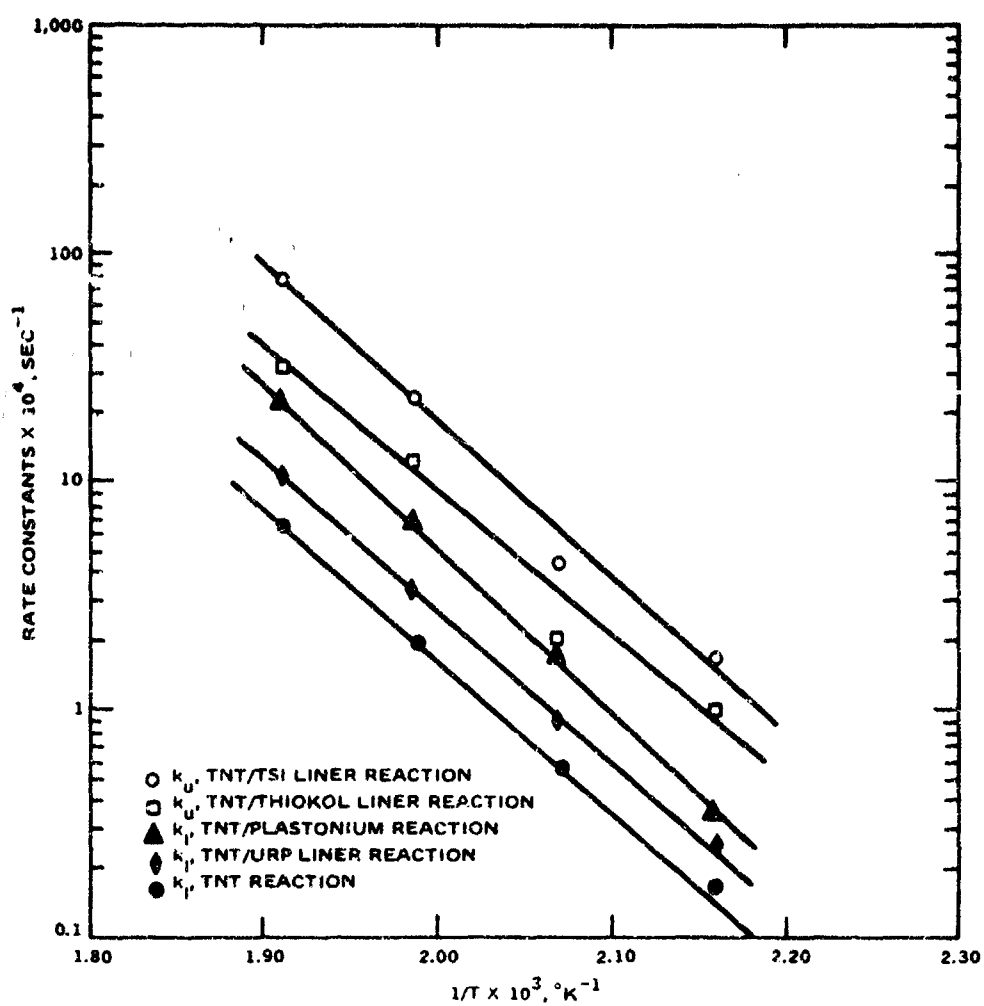


FIG. 18. Arrhenius Plots for the Decomposition of TNT in the Presence of Various Materials.

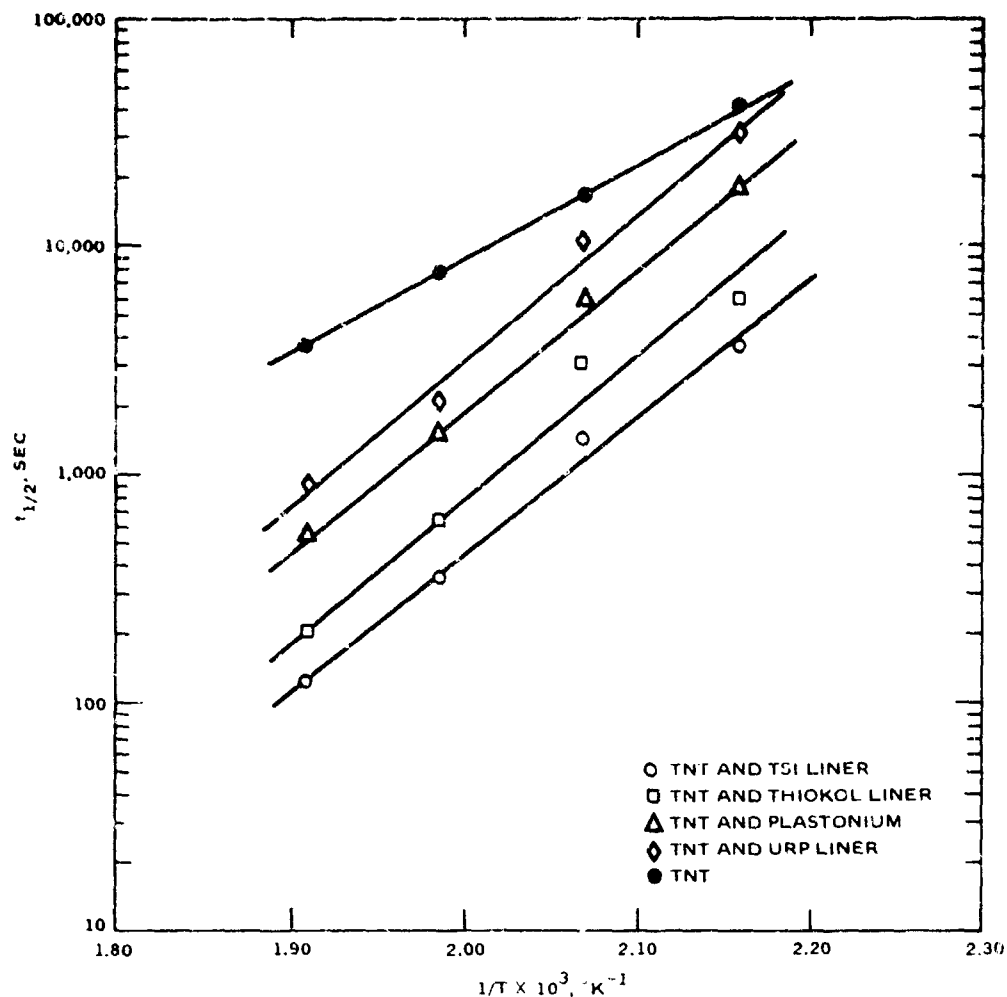


FIG. 19. Half-Life Plots for the Decomposition of TNT in the Presence of Various Materials.

TABLE 10. Rate Constants (k_d) for the Decomposition of TNT in the Presence of TSI and Thiokol Liners.

Temperature, °C	$k_d \times 10^4, \text{sec}^{-1}$	
	Additive	
	TSI Liner	Thiokol Liner
250	76.3	31.3
230	22.1	12.1
210	4.37	2.07
190	1.63	0.974

TABLE 11. Rate Constants (k_d)^a for the Decomposition of TNT in the Presence of Plastonium and URP Liners.

Temperature, °C	$k_d \times 10^4, \text{sec}^{-1}$		
	Additive		
	None	Plastonium ^b Liner	URP Liner
250	6.28	22.4	10.1
230	1.95	6.58	3.25
210	0.551	1.69	0.307
190	0.161	0.332	0.257

^a The rate constants approximately describe the region $0.30 < \phi < 0.85$.

^b The rate constants for the Plastonium Liner are based on data corrected for an initial evolution of water.

TABLE 12. Rate Constants (k_d) for the Decomposition of TNT in the Presence of Plastonium and URP Liners.

Temperature, °C	$k_d \times 10^4, \text{sec}^{-1}$		
	Additive		
	None	Plastonium ^a Liner	URP Liner
250	5.68	22.6	9.06
230	1.39	7.86	3.99
210	0.613	2.55	0.821
190	0.163	0.436	0.235

^a The rate constants for the Plastonium Liner are based on data corrected for an initial evolution of water.

TABLE 13. Half-Lives ($t_{1/2}$) for the Decomposition of TNT in the Presence of TSI, Thiokol, Plastonium, and URP Liners.

Temperature, °C	$t_{1/2}, \text{sec}^a$				
	Additive				
	None	TSI Liner	Thiokol Liner	Plastonium ^b Liner	URP Liner
250	3,978	126	210	553	927
230	8,664	360	642	1,566	2,124
210	16,180	1,452	3,180	5,628	10,560
190	44,340	3,720	5,940	18,240	32,380

^a The reported half-lives are uncorrected for a warm-up period of about 30 seconds.

^b The half-lives for the Plastonium Liner are based on data corrected for an initial evolution of water.

DISCUSSION

SUMMARY

This study investigated the behavior of two explosives in the presence of eight (counting the samples of Unique Liner as distinct) different materials. Results ranged from fairly promising inertness to three highly reactive and possibly dangerous systems. A quality control problem was identified in one case. Unusual decomposition features--for instance, the evolution of water--appeared in others. Under the circumstances, generalizations are difficult. Perhaps, in fact, the only really useful generalization would be a repetition of a previous statement (Ref. 8) to the effect that hot explosives, because of their inherent nature, may be expected to react with a wide variety of materials which might be employed as cavity liners. Previous reports stressed the possibility that such reactions may have serious consequences from the standpoint of ordnance safety and, in particular, the problem of munitions exposed to a fire.

Of the systems under study, Unique Liner sets a particularly difficult problem. The initial sample (Hard Unique Liner)--while certainly not inert--did not show an unusual reactivity with TNT and had fairly mild effects on Composition B. On the other hand, the second sample (Soft Unique Liner) must rate as one of the worst materials studied to date. The general accelerations, although obvious, are less alarming than the onset of unusually complete, violent reactions at the notably low temperature of 210°C. Subsequent verbal information⁵ indicated that due to a production error the sample was deficient in catalyst, an explanation in harmony with the very slow cure. At the least, then, a problem in quality control would seem to have been identified. (In fairness, it should be noted that the Unique Liner was the only case in which two independent samples were tested.) Investigations by J. M. Pakulak of NWC have indicated liner instability and an incompatibility of one of the components with explosives. The latter has also been reported in a Rocketdyne briefing (Ref. 3). All of this would seem to raise very serious questions as to the advisability of placing Unique Liner in intimate contact with explosives, particularly in munitions which may at some time be exposed to excessive heat.

Initial efforts to understand the difficulties with Unique Liner were handicapped by a total lack of knowledge of the chemical nature of the constituents. The components were identified only as "A" and "B," with even the most general information as to their nature undivulged. Subsequent information indicated that one of the ingredients was a tertiary amine. The red colors noted with TNT (Ref. 3) may therefore reasonably be attributed to a charge transfer complex of the amine and TNT. Some few further comments on the general problems raised by the normally unknown chemistry of the liners will be deferred to the closing section of this report.

Essex Liner and Tuffseal produced very serious accelerations. The Essex Liner had the greater effect on Composition B. Tuffseal produced the more serious accelerations with TNT. Moreover, Tuffseal had the worrisome property of producing pressure bursts at relatively low temperatures. Tuffseal is based on a petroleum asphalt, and asphalts generally accelerate the decomposition of explosives. However, the Tuffseal effects seem particularly severe, suggesting that some of the other constituents enter into the reaction.

TSI Liner produced accelerations roughly comparable to those caused by asphalts. Thiokol Liner had lesser but still noticeable effects. The Thiokol Liner withstood the temperatures of these tests without melting. It thus might be expected to have the advantage of remaining on the walls of the

⁵ J. M. Pakulak, private communication.

bomb and exerting its insulating effects for considerably longer times than would a low melting material. The TSI Liner melts at the reasonably high—higher than most asphalts—temperature of 150°C.

Following the initial evolution of water, Plastonium Liner had minor effects on Composition B and more noticeable, but still relatively mild, effects on TNT. However, the release of water was a very real feature of the sample we studied. Unfortunately, it is difficult to directly relate the observations in our vacuum line—where escape of water vapor (and other products) from the explosive was impeded only by 640 torr of nitrogen—with the interior of a bomb where confinement under increasing pressures would be expected.

The least reactive material under the conditions of these studies was URP Liner. The accelerations were minor with Composition B and less than those produced by the other candidates with TNT. The fact that the liner melts may limit its usefulness—at least from the standpoint of cook-off—since melting and mixing with molten TNT could bring explosive into contact with the hot metal bomb casing at relatively early times.

GENERAL COMMENTS

Previous reports discussed the possible practical implications of our results. These arguments will not be repeated. Nevertheless, a few summary comments may be useful. Our tests are deliberately taken to temperatures where reactions become rapid and the initiation of violent decomposition is imminent. It is entirely reasonable to assume that the processes under study in such regimes will be closely related to those initiating explosions in munitions exposed to a fire. Conventional low-temperature surveillance tests may—if much care is used in interpretation—provide useful information relative to long-term storage of munitions in magazines or in the field. They are far less likely to yield data directly pertinent to short-term exposure of munitions to fires.

It is obviously difficult to relate some aspects of our investigations to the far more complex system of a bomb (or shell) in a fire. However, it is worth noting that our tests use small samples in a clean system with the products permitted to escape from the vicinity of the explosive. It seems very probable that the reactivities of large masses of explosive under the confinement of a bomb or shell will be enhanced rather than reduced relative to those observed in our system.

Heat flow through the bomb casing and whatever insulation is available will certainly constitute the major factor in determining the time to cook-off. Nevertheless, the chemical reaction of the explosive (and any contaminants present) will ultimately initiate the explosion. It is entirely reasonable to assume that the more reactive the explosive system, the earlier explosion will occur. Differences in the reaction times of various explosive systems may well constitute a minor part of the total time to cook-off. They may also be entirely critical in practical circumstances where a very few seconds may be the difference between a damaging explosion and the successful cooling off of a threatened bomb. We have argued (Ref. 1) that this is likely to be particularly apparent from the standpoint of the firefighters attempting to reach the bomb.

In previous papers we have illustrated the effects of additives on the reactions of explosives by contrasting the temperatures necessary to achieve some particular half-life in the presence or absence of a foreign material. There seems little point in repeating the arguments in detail. Simple inspection of Fig. 5, 10, 14, and 19 will reveal temperature differences ranging from a few degrees for some Composition B systems to many tens of degrees for TNT-liner reactions. Such comparisons suggest, rather strongly, that the accelerations observed in these studies may have very real significance in practical systems. In the present investigation, the appearance of violent reactions at relatively low

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temperatures is an even more emphatic demonstration of the same point.

Experience to date has indicated that most liner candidates melt at relatively low temperatures. Melting probably reduces or eliminates their value as insulators, since convection may then carry a slurry of molten TNT, liner, and other explosive ingredients (RDX in the case of Composition B) into intimate contact with the hot metal casing. Presumably, explosion will follow shortly thereafter. It therefore seemed reasonable (Ref. 8) to suggest that efforts be directed toward developing a chemically inert liner which will remain on the walls of the bomb and thus provide a strong, rather than a weak, link in the casing/liner/explosive system.

Comments have been made here and in previous reports regarding the uncertain or entirely unknown chemical composition of the reactants. Often this was inherent in the nature of the material. For instance, asphalts and many waxes are complex mixtures of natural products with doubtful lot-to-lot reproducibility. With some proprietary materials the situation may worsen to the extent that even the classes of compounds are unknown. The disadvantages are obvious.

This investigation, for instance, has yielded detailed data on the reaction rates of a large number of explosive/additive systems. It has provided information on specific liner candidates, given some feeling for the behavior of explosives in contact with foreign materials, and reinforced the supposition that a very wide range of substances would react with hot explosives. Unfortunately, it has added little to our fundamental knowledge of the reactions of explosives. When the reactants are unknown, it is difficult to make sense of the chemistry.

Similarly, the behavior of new liner candidates cannot be predicted if their composition is unknown. This forces a tedious and expensive investigation of each new additive as it appears on the scene. A more sensible approach would build up a body of knowledge on the behavior of explosives in the presence of various, well-defined compounds and substances. This would permit a preliminary screening of candidates of known composition. Thereafter, efficient investigations--since they would be founded on reasonable chemical knowledge--would provide necessary checks and evaluate new formulations. In so doing, they in turn would add to the understanding of explosive/additive reactions.

Turning specifically to the problem of munitions: The goal, certainly, is the provision of ordnance which will be as safe as possible during manufacture, during storage in either magazines or the field, and during severe transient conditions such as would exist during exposure to a fire. During manufacture the explosive/liner system is exposed to heat--the temperatures necessary for casting--with a consequent possibility of significant chemical reaction. This may be microscopic, but far less than a milligram of material can be the source at some future time of the hot spot that initiates a detonation of hundreds of pounds of explosive. Following manufacture the bomb may be stored for years or decades under various conditions ranging from arctic cold to severe diurnal temperature cycling during field storage in the tropics or deserts. Thus very slow but potentially dangerous alterations in the liner/explosive systems become of clear importance. Finally, the bomb may be exposed to unintended hazards such as fire. This, it can be sensibly argued, should be thought of as an anticipated rather than an unexpected event. Munitions are designed for employment in a hazardous environment, with an enemy striving to increase the hazards still further. This being so, sooner or later some munitions are going to be exposed to a fire.

Presumably, in evaluating a given liner or sealant for application in direct contact with explosives, its behavior under all of the above conditions should be anticipated. Such predictions would be difficult even given a certain knowledge of the purity and composition of the explosive/liner system. It is totally absurd to suggest that they could even be attempted for unknown reactants. Surely, therefore, it would be sensible and proper to require a complete knowledge of all of the chemicals going into such a hazardous system as a bomb or shell.

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13. ABSTRACT The isothermal decompositions of Composition B and TNT were probed in the presence of a variety of materials under consideration for employment in ordnance as cavity liners or sealants. These materials included products of Essex Chemical Corp.; H. H. Robertson Co. (Tuffseal); Unique Products, Inc.; Insulation Systems Inc.; Thermal Systems, Inc.; Thiokol Chemical Corp.; and United Resin Products, Inc. The kinetic features of the reactions were established over wide temperature ranges, including conditions such that the reactions were fast and explosion imminent. In general, accelerations in the decomposition rates were observed. In a few cases these accelerations were extreme.			

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1a. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Thermal Decomposition of Explosives						
Composition B						
TNT						
Cavity Liners						
Sealants						
Cook-Off						

ABSTRACT CARD

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